PRELIMINARY EXPERIMENTAL RESULTS OF GAS RECYCLING SUBSYSTEMS EXCEPT CARBON DIOXIDE CONCENTRATION

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### ABSTRACT

Oxygen concentration and separation is an essential factor for air recycling in a CELSS. Furthermore, if the value of the plant assimilatory quotient is not coincident with that of the animal respiratory quotient, the recovery of  $\rm O_2$  from the concentrated  $\rm CO_2$  through chemical methods will become necessary to balance the gas contents in a CELSS. Therefore, oxygen concentration and separation equipment using Salcomine and  $\rm O_2$  recovery equipment, such as Sabatier and Bosch reactors, were experimentally developed and tested.

### 1. BASIC CONSIDERATION ON GAS RECYCLING

Fundamental functions of gas recycling in CELSS, as shown in Fig. 1. 1, are to separate each component, such as  $O_2$ ,  $CO_2$ , and  $N_2$  gases, and to concentrate and store each gas in order to supply appropriate gas concentration for human beings, plants and algae.

## 2. OXYGEN RECOVERY FROM CONCENTRATED CARBON DIOXIDE

Many research and development efforts have been conducted for the recovery of oxygen from concentrated  $\mathrm{CO}_2$  by the catalytic hydrogenization process. The Bosch reaction utilizes an iron catalyst and produces carbon (C) and water ( $\mathrm{H}_2\mathrm{O}$ ) with CO as an intermediate product /1/. Product water is further electrolyzed to recover  $\mathrm{O}_2$  for animal respiration and  $\mathrm{H}_2$  for subsequent hydrogenization. The Sabatier reaction utilizes a ruthenium (Ru) catalyst and produces methane and water /2/. An additional  $\mathrm{CH}_4$  cracking process providing C and  $\mathrm{H}_2$  is necessary to make the Sabatier reaction comparable to the Bosch reaction /3/.

Mitsubishi Heavy Industries, Ltd. (MHI) has been involved in CELSS research for several years /4/ under the direction of the National Aerospace Laboratory and conducted the experimental program to evaluate the basic characteristics and performance of the two oxygen recovery processes.

# 2.1 Once Through Tests

In order to determine the basic characteristics of each reaction, once through tests of  $\rm CO_2$  reaction with  $\rm H_2$  on the catalyst are performed. Fig. 2.1 shows the schematic of the test set up.

Bosch Reaction rate of  $\mathrm{CO}_2$  vs. reaction temperature in the first Bosch reaction, as well as the reaction rate of  $\mathrm{CO}$  vs. temperature in the second Bosch reaction. The different nature of the two reaction rate curves suggests a phased process concept using two reactors operating at different temperature ranges to obtain higher performance in the Bosch  $\mathrm{CO}_2$  cracking process.

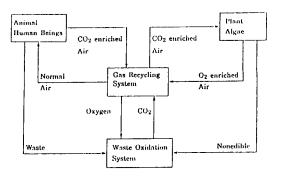


Fig. 1.1 Fundamental function of Gas Recycling System

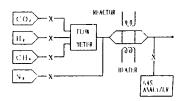


Fig. 2.1 Bosch/Sabatier
Once Through Test Setup

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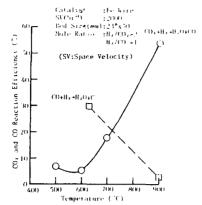


Fig. 2.2 Reaction Efficiency vs Temperature of Bosch Reaction

<u>Sabatier Reaction</u> Fig. 2.3 shows the reaction rate of  $CO_2$  vs mole ratio of  $H_2/CO_2$  in the feed gas mixture of the first Sabatier reaction. Reaction rate of more than 99 % is achieved with a little bit higher ratio (4.5) than the stoichiometric value. As for the second Sabatier reaction, reaction rate of  $CH_4$  vs. time after start of the reaction is presented in Fig. 2.4. Very rapid degradation is observed for the catalytic reactions using Pt or Ni, while a steady and much higher conversion efficiency is observed in pyrolytic reaction on silica wool filler.

# 2.2 Recycle Test

A Recycle Test Apparatus was prepared based on the once through test results as shown in Fig. 2.5.

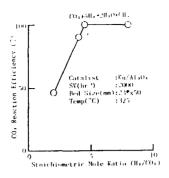


Fig. 2.3  $CO_2$  Reaction Efficiency vs  $H_2/CO_2$  Mole Ratio for Sabatier First Reaction

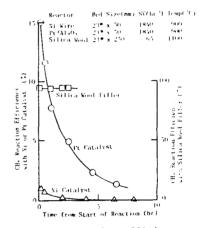


Fig. 2.4 CH<sub>4</sub> Creacking Efficiency vs Time for Sabatier Second Reaction

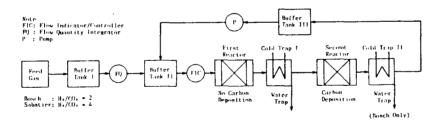


Fig. 2.5 Bosch/Sabatier Recycle Test Setup

The supply rate of mixture gas is shown in Fig. 2.6. The amounts of processed  $\mathrm{CO}_2$  are 0.15 kg/day and 0.42 kg/day for Bosch and Sabatier, respectively. Energy required for the above processes were estimated and the Sabatier process showed several times less energy use than Bosch process. The character of deposit carbon in the Bosch process was a loose powder form, while a hard solid block was obtained from the Sabatier reactor. This fact has an important meaning for maintainance operations to periodically extract carbon on orbit.

Thus, the  ${\rm O}_2$  recovery system with Sabatier methane cracking shows a good possibility for application in CELSS and Space Station.

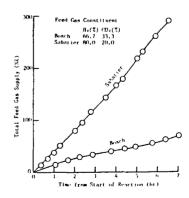


Fig. 2.6 Feed gas Supply Rate in Bosch/Sabatier Recycle Process

#### 3. OXYGEN SEPARATION SYSTEM USING SALCOMINE

## 3.1 Functions of Oxygen Separation System

Kawasaki Heavy Industries Ltd. (KIII) has been involved in CELSS research under contract with the National Aerospace Laboratory (NAL) /5/.

Fig. 3.1 shows the function of  $O_2$  separation and concentration system, concerning the  $CO_2$  and  $N_2$  separation and concentration. In the Fig. 3.1, the inlet gas is mixture of  $O_2$ ,  $CO_2$  and  $N_2$ . At the  $CO_2$  concentrater,  $CO_2$  is separated and concentrated, and stored in the  $CO_2$  gas bottle. At the  $O_2$  concentrater,  $O_2$  is separated and concentrated, and stored in the  $O_2$  gas bottle. The residual

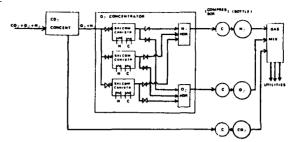


Fig. 3.1 Function of Oxygen Separation and Concentration System

is  $N_2$  gas and this is stored in the  $N_2$  gas bottle. Thus, inlet gas is separated into three (3) components,  $CO_2$ ,  $O_2$ , and  $N_2$  having high purities.

There are many methods for  $CO_2$  separation and concentration. The studies reported here are focused on the  $O_2$  separation and concentration system using Salcomine. Salcomine absorbs the  $O_2$  under normal temperature (lower than about 40°C) and desorbs the  $O_2$  under high temperature (higher than about 80°C). So, when the inlet gas is introduced into the Salcomine canister, the included  $O_2$  is absorbed by Salcomine. After the Salcomine absorbs the  $O_2$ , the Salcomine canister is heated and Salcomine desorbs  $O_2$  of high purity. In the Fig. 3.1, there are three (3) canisters and each canister is operated in absorbing, desorbing and pre-cooling modes respectively. Thus, continuous  $O_2$  separating operation can be carried out by the cyclic exchange of these operation modes.

### 3.2 SALCOMINE

Fig. 3.2 shows the structural formula of the Salcomine and its  $O_2$  absorption and desorption. There are some studies on the application of Salcomine for  $O_2$  concentration. Matsuda /6/ carried out the experimental study on the  $O_2$  absorbing and desorbing characteristics and their variations under repeated reaction of the Salcomine, as a part of the development of Diffusive Atmosphere Control System (DACS, a kind of artificial gill), which extract  $O_2$  from sea water dissolving oxygen.

The Present study is based on these results and is designed to get data to determine the optimum  $O_2$  separating and concentrating system for CELSS. In this study,  $O_2$  absorbing and desorbing performance tests of Salcomine within the canister are carried out to get the data to design a compact, light weight and lower energy consuming system.

# 3.3 Oxygen Abosrbing and Desorbing Test of SALCOMINE

3.3.1 Test of SALCOMINE Proper. In Fig. 3.3(a), oxygen containing nitrogen gas is introduced into the equipment and Salcomine absorbs the  $O_2$ , and increases its weight. And then the Salcomine is heated and the Salcomine decrease its weight desorbing  $O_2$ . From the weight change of the Salcomine, the  $O_2$  absorbed and desorbed is measured. Test results are shown in Fig. 3.3(b).  $O_2$  absorbing and desorbing capacity of Salcomine is expressed with the percentage of oxygen weight to the Salcomine weight (wt%). From Fig. 3.3(b),  $O_2$  absorbing capacity is about 4.0 wt% for 20% of  $O_2$  concentration.

3.3.2 Test of SALCOMINE within Canister. In Fig. 3.4,  $O_2$  containing air is drawn by the air pump and introduced into the Salcomine canister. In the canister,  $O_2$  in the air is absorbed by Salcomine.

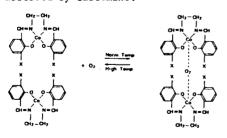
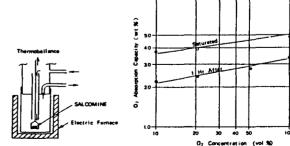


Fig. 3.2 Oxygen Absorb and Desorb
Reaction by SALCOMINE



(a) Test Equipment (b) Test Results
Fig. 3.3 Characteristic Test of SALCOMINE

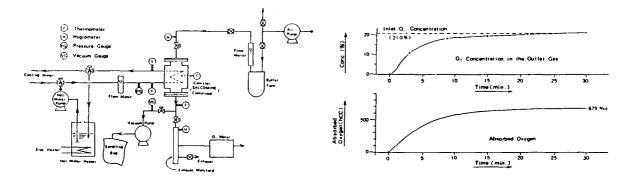


Fig. 3.4 Diagram of Canister Contained SALCOMINE Performance Test

Fig. 3.5 Results of Canister Contained SALCOMINE Performance Test

And at the exhaust gas manifold,  $O_2$  concentration is measured. During this time, The Salcomine canister is cooled. For the desorbing test, the canister is heated by hot water (90 °C) and drawn by the vacuum pump.  $O_2$  desorbed from the Salcomine is collected into the sampling bag and analysed.

Fig. 3.5 shows the results of the test. Initially,  $O_2$  concentration in the outlet air is almost  $O_0^*$  because all  $O_2$  in the air is absorbed. After a few minuits, the break-through occurs and  $O_2$  concentration rises, and reaches to 21% which is equal to that of the inlet air. From Fig. 3.6, absorbed  $O_2$  is 675CC and this corresponds to 2.7 wt% of  $O_2$  absorbing capacity. On the start of  $O_2$  desorbing test, the vacuum pump is operated to purge the air inside the test equipment. The canister is then heated. And is drawn by the vacuum pump. The drawn gas is measured 710 CC, and  $O_2$  concentration in this gas is measured 91.4%. So, desorbed  $O_2$  becomes 649 CC.

### 4. CONCLUSION

As a part of CELSS Gas Recycling System Development, preliminary experimental studies were conducted for the processes of  $O_2$  recorvery from concentrated  $CO_2$  and of  $O_2$  separation and concentration from exhaled gas of animal, plant etc.

Sabatier process for the regenerable  $O_2$  recovery from  $CO_2$  shows a good possibility compared with Bosch process in its higher efficiency of  $O_2$  recovery and easier handling of product carbon. From the  $O_2$  separation and concentration tests using Salcomine, oxygen absorbing and desorbing characteristic data are obtained as the fundamental design data.

The above results can be used in the planning of a complete and stable gas recycling system required in CELSS. We hope to continue our research and development effort for the expansion of the human frontier in Space.

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